

# APPLICATION OF THE MEDEC PROCESS TO TREAT FERMI-1 SODIUM-BONDED SPENT NUCLEAR FUEL

Karen L. Toews  
Steven D. Herrmann  
David A. Sell  
Richard H. Rigg  
Robert G. Pahl  
Argonne National Laboratory  
P.O. Box 2528  
Idaho Falls, ID 83404

## ABSTRACT

Argonne National Laboratory is currently investigating the feasibility of and developing a life-cycle cost estimate for the treatment and disposal of 34 metric tons of sodium-bonded Fermi-1 blanket fuel using the Melt Drain Evaporate Carbonate (MEDEC) process. The scope of this work includes fuel characterization and specification, dose rate calculations, flow sheet development, process demonstration tests using unirradiated and irradiated fuel, equipment layouts in two candidate facilities, development of dynamic process models, and preparation of a low-uncertainty life-cycle cost estimate. A description of the process flow sheet, facility operations, and the results of the unirradiated testing activities are presented. A brief description of irradiated testing activities that are currently underway is also given.

## INTRODUCTION

The Department of Energy currently manages 60 metric tons of sodium-bonded spent nuclear fuel, including fuel from the Experimental Breeder Reactor-II (EBR-II), Enrico Fermi Atomic Power Plant (Fermi-1), and Fast Flux Test Facility (FFTF) reactors. The Record of

Decision for the Final Environmental Impact Statement (EIS) for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel was issued in September 2000, and stated that the EBR-II and FFTF fuel would be treated electrometallurgically at Argonne National Laboratory-West (ANL-W). However, the treatment decision for the remaining 34 metric tons of Fermi-1 blanket spent nuclear fuel was deferred in order to "validate the cost of using treatment alternatives." [1] Alternative treatment approaches may be economically and technically feasible for the Fermi blanket fuel due to its significantly lower plutonium content and correspondingly less stringent safeguards and security requirements. Several alternatives to electrometallurgical treatment including direct disposal and the melt and dilute technology will require sodium removal. The Melt-Drain-Evaporate-Carbonate (MEDEC) treatment process is one method by which the bond sodium may be removed from fuel elements and the sodium stabilized.

The aim of the present study is to examine the issues associated with treatment of the fuel at ANL-W and subsequent packaging of the treated fuel. Results of flow sheet development, facility selection, and glovebox and hot cell testing activities using unirradiated Fermi fuel and irradiated EBR-II fuel are presented.

## FERMI-1 BLANKET FUEL DESCRIPTION

The Fermi-1 blanket fuel consists of cylindrical rods of uranium housed within stainless steel 304 cladding. The annular region between the fuel and cladding contains metallic sodium that serves as a thermal bond. Two types of blanket

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fuel configurations were used in the Fermi reactor: axial and radial blanket fuel. Both fuel configurations consist of 97% depleted uranium (0.35%  $^{235}\text{U}$ ) alloyed with 3% molybdenum. The radial elements are 71.5 inch (181.6 cm) long and contain 2.2 kg heavy metal (HM) and approximately 23 g of metallic sodium. Each radial element contains four depleted uranium slugs that are 14 inches (35.6 cm) long and one fuel slug that is 5.75 inches (14.6 cm) long. Thus, the total length of the radial element fuel column is 61.75 inches. The region above the fuel column, known as the fuel plenum, contains argon fill gas. The sodium metal extends about 2.5 inches into the plenum region. The 17.5 inch (43.2 cm) long axial fuel contains a single 14 inch (35.6 cm) long slug of depleted uranium. The annular region of the fuel contains approximately 6 g of metallic sodium. As with the radial blanket elements, sodium from the fuel column region extends a short distance (about 1.2 inches) into the plenum region of the axial element and the remainder of the plenum contains argon fill gas. These and other relevant fuel specifications are summarized in Table I.

**Table I: Nominal Characteristics of Fermi Radial and Axial Blanket Fuel Elements**

	RADIAL	AXIAL	NOTES
Cladding outer diameter, in	0.443	0.443	Ref. [2]
Cladding wall thickness, in	0.010	0.010	Ref. [2]
Sodium radial annulus thickness, in.	0.014	0.014	Ref. [2]
Fuel slug diameter, in.	0.395	0.395	Ref. [2]
Fuel column total length, in	61.75	14	Ref. [2]
"Excess" sodium height above fuel column, in.	2.5	1.2	Axial value per X-radiograph Radial, Ref. [2]
Fuel element length, in.	71.5	17.5	Ref. [2]
Uranium, total g per fuel element	2,211	501	Appendices A-3, A-1 of Ref. [3]
Sodium, g per fuel element	23.17	6.22	Calculated with nominal dimensions, 0.967 g/cc

The axial fuel elements were configured into assemblies that contained 16 fuel elements arranged in a 4x4 array. The assemblies were located above and below the reactor core. Radial blanket assemblies were located around the periphery of the reactor; each assembly

contained 25 blanket elements in a 5x5 array. The inventory of Fermi fuel includes 403 axial and 559 radial blanket assemblies.

Fermi reactor core physics calculations for individual blanket assemblies were obtained from a Detroit Edison internal report. [4] A computer code, FERMUP, was used to calculate the data presented in the report. The calculated average burn-up for both axial and radial assemblies is 0.0017%. The highest burn-up assembly was a factor of 10 higher than the average burn-up. The average plutonium content in the axial and radial blanket assemblies is 3.6 g and 10 g, respectively. The maximum plutonium content present in the axial and radial assemblies is 9.2g and 39.3 g, respectively.

Calculations have been completed that estimate the dose rates associated with axial and radial Fermi-1 blanket fuel. These calculations are a critical component in estimating the fuel treatment costs. The radiation field calculations will be used to determine facility, enclosure and handling options for MEDEC treatment of the Fermi fuel at Argonne National Laboratory-West (ANL-W). The details of these calculations are given elsewhere. [5]

Dose rates were calculated using the QAD-FN code. The calculated dose rate for an average axial blanket assembly is 1.5 rem/hr at contact and 20 mrem/hr at 1 meter. The dose rate from the highest burn-up axial assembly is calculated to be 4.4 rem/hr at contact and 59 mrem/hr at 1 meter. Likewise, the calculated dose rate for an average radial blanket assembly is 2.7 rem/hr at contact and 108 mrem/hr at 1 meter. The dose rate for the highest burn-up radial assembly is expected to be 20 rem/hr at contact and 800 mrem/hr at 1 meter. For consistency, the dose rates are reported as of January 2000.

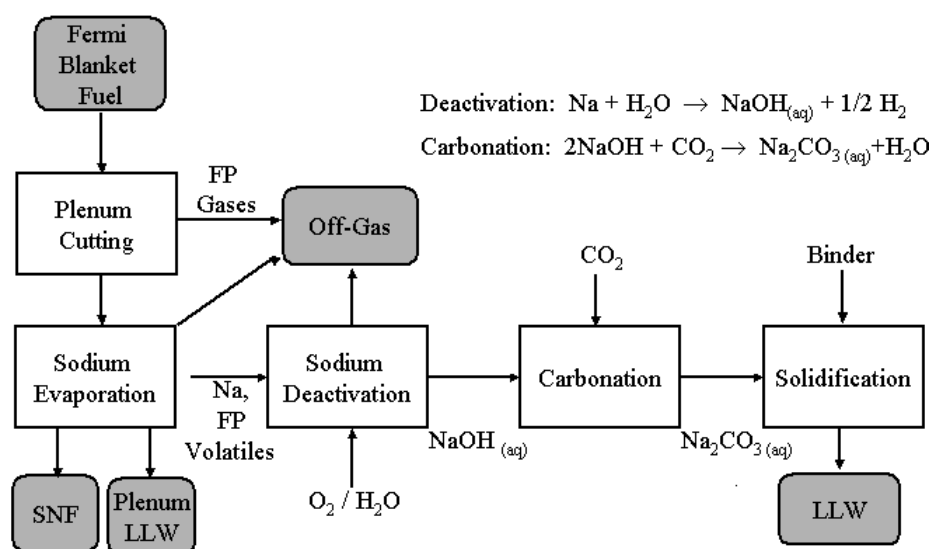
## FLOW SHEET DEVELOPMENT

A flow sheet for MEDEC treatment operations has been developed and is given in Fig. 1. This flow sheet includes the following unit operations: plenum cutting, sodium evaporation, sodium deactivation, carbonation, and solidification.

First, the sodium-bonded fuel assemblies are transferred into an inert atmosphere shielded glovebox or hot cell where the assemblies are

sectioned through the plenum region to expose the sodium within the cladding. The cut assemblies are then placed within an evaporation vessel and inserted into a furnace. The furnace temperature is raised to approximately 650° C and a vacuum is applied to the vessel. This causes the bond sodium to melt and vaporize. The sodium is collected in a condenser. The cleaned fuel elements are removed from the evaporation vessel and placed in sealed canisters for interim storage and final disposal. The plenum sections are likewise treated and then are packaged for disposal as low-level waste (LLW).

**Figure 1: MEDEC flow sheet**



Next, the collected sodium is deactivated by injection into a chamber where it reacts with oxygen from the air and water to form aqueous sodium hydroxide. Carbon dioxide gas is subsequently bubbled through the hydroxide solution converting the sodium hydroxide to sodium carbonate. The aqueous sodium carbonate is solidified with a binder to comply with land disposal requirements prohibiting disposal of free liquids (10 CFR Part 61), and then is packaged for disposal. The classification of this waste is under evaluation, but is expected to be contact-handled LLW.

## FACILITY OPTIONS

Facility selection is a critical factor determining the cost of treatment operations at ANL-W. Due to the low dose rates of the Fermi fuel, two

treatment approaches are feasible. The first option involves performing treatment operations in a semi-remote fashion using a shielded glovebox located at ANL's Transient Test Reactor (TREAT) facility. The second treatment option entails performing MEDEC operations in a fully shielded hot cell housed within ANL's Hot Fuel Examination Facility (HFEF). The TREAT option has the advantage of lower facility overhead and is expected to require shorter process times. Unlike HFEF, the TREAT glovebox facility will house only a single project; thus conflicts associated with station and resource utilization will be minimized. Furthermore, the TREAT glovebox will be designed around the process resulting in an

optimized layout for maximum efficiency.

This should result in a higher throughput, and a shorter duration treatment campaign. However, the TREAT option will require more extensive design and fabrication efforts since both a shielded glovebox and fuel transfer tunnel will be required.

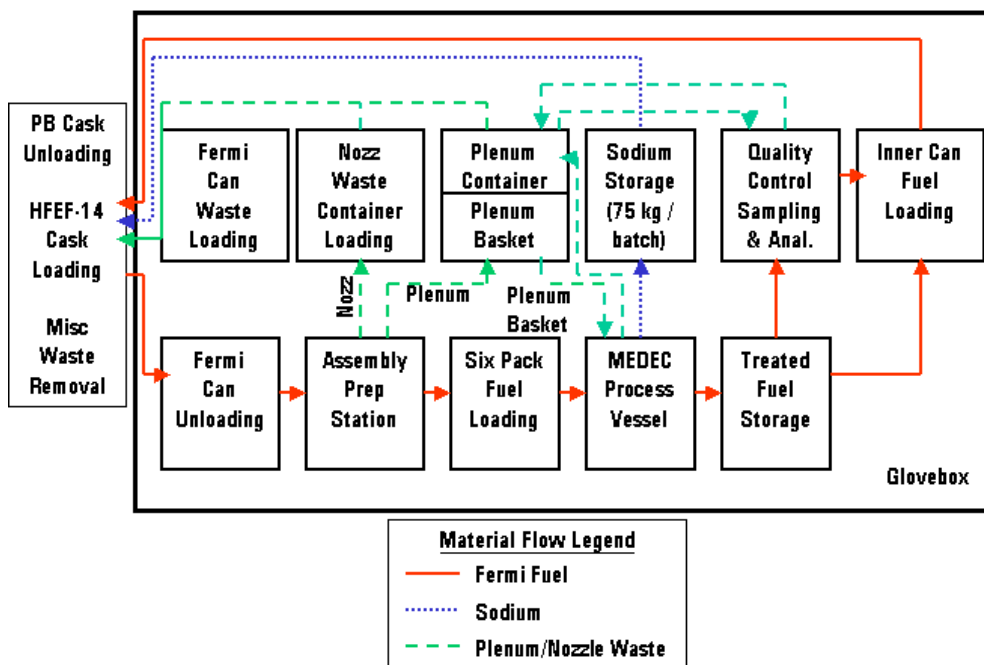
The HFEF option, in contrast, has the benefit of using an existing enclosure and cask tunnel, but is likely to

require lengthier handling operations, and more elaborate and costly fully remotized equipment. The facility overhead will also be higher and conflict with existing facility operations may be problematic. Both options are more explicitly explored below.

Material flow through the TREAT facility is expected to proceed as shown in Fig. 2. Fuel will be transferred into the glovebox via a below grade tunnel. The fuel will be staged and prepared for treatment by first removing the fuel assembly nozzle, installing lifting fixtures and then cutting through the plenum region of the fuel to expose the sodium. Next, an element retaining fixture will be installed to keep the elements in the assembly housing. The intact assembly will be inverted and placed into a basket, designed to hold 6 fuel assemblies. Once full, the basket will be transferred into the MEDEC vessel where sodium removal will

occur. The sodium will be collected in 30 gallon batches for subsequent deactivation in a separate facility. The fuel will be transferred to an inner storage canister with a capacity for either 20 radial or 60 axial blanket assemblies. The plenum pieces removed in the initial preparation operation will be collected and processed to remove residual sodium. Residual sodium analysis will be performed on both the fuel and plenum sections for quality control. Once the assemblies and plenum sections are certified to be free from sodium, they will be packaged and removed from the glovebox using an ANL HFEF-14 can. The plenum and other ancillary assembly hardware will be packaged for disposal as LLW, while the cleaned fuel will be stored at an interim storage facility pending its final disposition in a repository. The recovered sodium will be deactivated at ANL's Sodium Component Maintenance Shop (SCMS) using the procedure outlined in the flow sheet. Sodium will be processed in 30 gallon batches, and the resulting solidified sodium carbonate will be disposed of as LLW (pending verification by irradiated tests described below).

**Figure 2: Treat glovebox workstations**



### Layout and Material Flow

MEDEC operations in HFEF will proceed in a similar manner to those in TREAT with the exception of differences in the workstation

layout. The HFEF presently houses electrometallurgical waste processing operations associated with treatment of EBR-II fuel. The MEDEC equipment layout and processing operations have been designed to utilize existing hot cell equipment (e.g. cranes, cask tunnel, electromechanical manipulators) where possible, and to cause minimal interference with existing facility operations.

### MEDEC DEMONSTRATION TESTS

MEDEC demonstration tests were designed to answer several fundamental questions regarding process feasibility; specifically, these tests were aimed at determining necessary operating conditions, degree of sodium removal, and the co-extraction of cesium. Testing was initiated in two phases. Phase I testing involved performing MEDEC treatment on unirradiated Fermi blanket fuel to examine process feasibility and sodium removal efficiency. Phase II testing is currently underway and will involve MEDEC treatment of irradiated EBR-II fuel to ascertain the amount of fission product cesium transported during the sodium distillation process.

### Unirradiated Fermi Fuel Tests

Activities ranging from small scale testing to process scale operations have been performed at ANL-W to develop and optimize the MEDEC process. These tests have evaluated process conditions and rates, and the efficiency of sodium removal. A brief summary of previously performed testing activities is included since these tests were foundational to the

present study.

In the early 1980s, the MEDEC process was developed as a treatment method to remove sodium from unirradiated fuel elements and from

sodium-bearing wastes. [6] These initial tests were performed at different process conditions than are proposed for the present work, namely, evaporation was performed at 500 °C with a  $10^{-4}$ – $10^{-5}$  Torr vacuum. Vapor conductance tests were performed to evaluate the process applicability to the removal of sodium from crevices and “chimneys”, such as those found in cold traps and the annular region of fuel elements. These tests confirmed that the MEDEC process could remove sodium from chimneys 20.3 cm deep with a 0.127 mm gap width. Post-test specimens were visually examined for sodium and were chemically analyzed for residual sodium. Following these development tests a full-scale processing campaign was initiated wherein 1700 unirradiated EBR-II fuel elements were processed. [7] The sodium was completely removed from these elements. Finally, a full-size EBR-II cold trap was processed, again with excellent results. While the results of this test campaign were excellent, the installation, operation and maintenance of high vacuum equipment in a hot cell environment were a concern.

In an effort to avoid the installation of high vacuum pumps (e.g. cryogenic pumps or turbomolecular pumps) in a hot cell, a higher temperature evaporation step was proposed, thereby lessening the vacuum requirement. As mentioned above, present MEDEC treatment operations will be performed at 650 °C with a  $10^{-3}$  Torr vacuum, rather than the 500 °C /  $10^{-4}$ – $10^{-5}$  Torr conditions used in the aforementioned tests. To validate these operating conditions for sodium removal, a testing campaign was initiated using unirradiated Fermi fuel elements.

Both radial and axial unirradiated Fermi blanket fuels were utilized in the testing operations. As discussed previously, the radial elements are 71.5 inch (181.6 cm) long and contain approximately 23 g of metallic sodium. Each radial element contains four depleted uranium slugs that are 14 inch (35.6 cm) long and one fuel slug that is 5.75 inch (14.6 cm) long. The radial elements were prepared for testing by segmenting the elements at the fuel slug junctions, yielding 4 test specimens 14 inches (35.6 cm) in length and one test specimen (containing the shorter, 5.75 inch fuel slug) 15.5 inches (39.3 cm) in length. The 17.5 inch (43.2 cm) long axial fuel elements contain a single 14

inch (35.6 cm) long slug of depleted uranium and approximately 6 g of metallic sodium. The axial elements were prepared for testing by making a single cut through the plenum region of the fuel element.

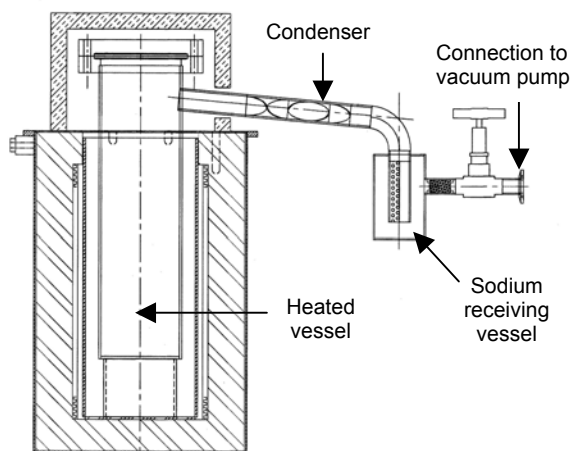
Following the segmenting operation, the Fermi fuel test material was processed at 650 °C and 200 mTorr for 4-6 hr in the apparatus shown in Fig. 3. The apparatus consisted of a lower heated test portion, a viewing port, and a condenser. The tests were performed in an existing inductively heated furnace and used a 600 L/min scroll-style dry vacuum pump. An in-line sodium sensor, consisting of a sodium hollow cathode lamp, a bandpass filter, and a photomultiplier tube was employed to monitor the evaporation process. This equipment was housed in an argon atmosphere glovebox with a maximum moisture and oxygen content of 100 mg/kg (100 ppm).

**Figure 3: MEDEC glovebox test apparatus**



A total of 6 tests were performed using two fuel segments per test. These tests utilized several different lengths of fuel and included test specimens with either one open end or two open ends through which the sodium could evaporate.





The aforementioned equipment has been designed and fabricated and is currently being qualified for hot cell operation. These qualification tests will verify that the equipment design is adequate for efficient sodium evaporation and collection.

## **PATH FORWARD**

Following the completion of the hot cell tests and the conceptual equipment layout for both the TREAT and HFEF facilities, a life-cycle cost estimate will be calculated for each facility option. This will include costs for shipping, receiving, treating and packaging the fuel and waste, and will include costs for interim and final disposal. Once these cost estimates are obtained, they will be submitted to the Department of Energy, along with a process feasibility report. This information, in conjunction with information given in the "Cost Study for Alternatives presented in the Draft EIS for the Treatment and Management of Sodium-Bonded Spent Nuclear Fuel," will be used to direct the final treatment decision for the Fermi blanket fuel. [8] Presently, the most mature technologies appropriate for treatment of sodium-bonded fuels include MEDEC treatment as described herein, and electrometallurgical treatment. As mentioned previously, electrometallurgical treatment is presently being used at ANL-W to treat 26 MT of sodium-bonded EBR-II and FFTF fuels. The Fermi fuel, and all other fuel currently stored in Idaho is legally required to be shipped out of the state by 2035 per the 1995 Idaho Settlement Agreement. [9]

## **ACKNOWLEDGEMENTS**

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